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Computer simulation, nuclear techniques and surface analysis

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This article is about computer simulation and surface analysis by nuclear techniques, which are non-destructive. The "energy method of analysis" for nuclear reactions is used. Energy spectra are computer simulated and compared with experimental data, giving target composition and concentration profile information. Details of prediction stages are given for thick flat target yields. Predictions are made for non-flat targets having asymmetric triangular surface contours. The method is successfully applied to depth profiling of ^{12}C and ^{18}O nuclei in thick targets, by deuteron (d,p) and proton (p,α) induced reactions, respectively.

Keywords: Thick/thin films, Surfaces, Non-destructive tests, Nuclear reactions, Computer simulation.

Simulación por ordenador, técnicas nucleares y análisis de superficies

Este artículo trata de simulación por ordenador y del análisis de superficies mediante técnicas nucleares, que son no destructivas. Se usa el "método de análisis en energía" para reacciones nucleares. Se simulan en ordenador espectros en energía que se comparan con datos experimentales, de lo que resulta la obtención de información sobre la composición y los perfiles de concentración de la muestra. Se dan detalles de las etapas de las predicciones de espectros para muestras espesas y planas. Se hacen predicciones para muestras no planas que tienen contornos superficiales triangulares asimétricos. Este método se aplica con éxito en el cálculo de perfiles en profundidad de núcleos de ^{12}C y de ^{18}O en muestras espesas a través de reacciones (d,p) y (p,α) inducidas por deuterones y protones, respectivamente.

Palabras clave: Películas espesas/finas, Superficies, Tests no destructivos, Reacciones nucleares, Simulación por ordenador.

1. INTRODUCTION

There is a broad range of surface analysis techniques which are, generally, complementary and provide target information for depths near the surface. Nuclear techniques, which are non-destructive, provide for analysis over a few microns close to the surface giving absolute values of concentrations of isotopes and elements. They have been applied in areas such as scientific, technologic, industry, arts and medicine, using MeV ion beams (1-6). Nuclear reactions permit tracing of isotopes with high sensitivities. We use ion-ion reactions and the energy analysis method. At a suitable energy of the incident ion beam, an energy spectrum is recorded of ions from the reaction, coming from several depths in the target. Such spectra are computationally predicted, giving target composition and concentration profile information (4-7). Elastic scattering is a particular and important case. A computer program has been developed in this context, mainly for flat targets (4-6). The non-flat target situation arises as an extension. For the $^{18}O(p,\alpha_0)^{15}N$ reaction, computer simulations are made for non-flat thick oxides having asymmetric surface contours. This permits spectral shapes to be compared, taking the flat target spectrum as a reference. Applications of the method are made to thick oxides obtained by high temperature oxidation of steel samples. In addition, the $^{12}C(d,p_0)^{13}C$ reaction is applied to ^{12}C depth profiling for the thick target case. The rest of the paper includes: the experimental details and samples; computer simulation; results and discussion; conclusions.

2. EXPERIMENTAL DETAILS AND SAMPLES

The experimental arrangement has been given (6). Ion detection from nuclear reactions in the samples was at a laboratory angle θ_L of 165º. Spectra were acquired as counts per channel versus channel number. Following energy calibration of these spectra, spectral yields as counts per unit energy versus energy were obtained.

We used the following samples as targets for acquisition of charged particle spectra: 1)A thick, high purity, sample of *pyrolitic graphite*, labelled *S1*; this sample was made by Union Carbide, by cracking CH_4 at 2200 ºC and depositing onto a graphite substrate; the target had a flat surface, as verified by scanning electron microscopy; 2) an austenitic steel (20/25/Nb steel) sample, labelled *S2*, which was oxidized at 650 ^{O}C in C $^{18}O_2$ gas for 121 hours; the oxide was reasonably flat, as verified through scanning electron microscopy; from weight gain measurements an oxide thickness of about 4.2 μm was expected; a thickness of 3.30 μm had been determined by the "resonance method" for the 1.763 MeV resonance of the $^{18}O(p,\alpha_0)^{15}N$ reaction (8).

3. COMPUTER SIMULATION

A large scale computational procedure has been developed for simulation of energy spectra of charged particles from nuclear reactions and, as a particular and important case, elastic scattering from targets under ionic bombardment (4-6). Computer simulated spectra are compared to experimental data through a chi-square procedure, to evaluate the goodness of fit. Through variation of target parameters, so as to fit experimental data, target composition and concentration profiles are obtained.

Computer simulation, in building spectral predictions, mainly accounts for: target parameters, such as composition and concentration profiles; energy spread of the incident ion beam; geometric factors and target rotation; stopping power; differential cross section; energy straggling; detector resolution. An option permits calculation of effects from: small forward angle multiple scattering; incident beam size and angular divergence; detector angular aperture. Gaussian straggling is considered, based on Bohr theory (9). Lindhard-Scharff theory (10) is the main option. X_i is a target or target layer thickness parameter. In building a basic spectrum, with ingoing ions penetrating the target and outcoming ions travelling towards the detector, we calculate in short: mean ingoing energies and ingoing energy distributions; depth dependence of the yield; mean energies and energy distributions from nuclear reaction events; mean outcoming energies and outcoming energy distributions; dependence of the yield on mean outcoming energy; dependence of the yield on outcoming energy; dependence of the yield on detection system resolution, giving final yield versus energy.

Fig. 1 represents a nuclear reaction experiment using a non-flat target. An incident ion beam of ions *a* is considered with a small profile of area S, perpendicular to the direction of incidence, and uniform probability distribution function $j(y,z)=j_0$ of the beam current density, such that

$$\iint_S j(y, z)\,dydz = 1 \tag{1}$$

The surface of the target consists of points whose coordinates are y, z, $h=\phi(y,z)$. The energy spectrum of ions *b* from a reaction A(*a*,*b*)B in the target is composed of spectra from elemental target volumes whose cross section areas are dS=dy dz in the plane perpendicular to the direction of incidence, i.e.

$$\mathrm{spect}_b^{(2)}(E_{bf}, S) = j_0 \iint_S \mathrm{spect}_b^{(2)}(E_{bf}, y, z)\,dydz \tag{2}$$

The function under integration explicitly includes the dependence on y and z. The dependence on other relevant quantities and on surface topography is included through the dependence on E_{bf}, which represents observed energy at the detection system. That function is the complete energy spectrum of ions *b* from a target elemental volume corresponding to a beam area dS. We consider a beam of area $S=4L^2$, 2L being the transversal dimension. Predictions are made, through division of the area S into N elemental areas $\Delta S_i=\Delta y_i\ \Delta z_i$ for i=1, N. In this way, the target is divided into N elemental volumes of cross section ΔS_i. In every plane of constant y, for ions entering the target surface at coordinates y_i, z_i, h_i the distribution of outcoming versus ingoing path lengths is calculated. This distribution depends, for a detection angle θ_L, on the coordinates of the entrance point and on surface topography. Final yield versus energy arises in a way similar to the flat target case. In Fig. 2, parameterization is given for a flat target having an asymmetric surface contour. The surface has triangular grooves along the y axis. The contour is the same for all planes of constant y. The main parameters shown are H_0, Z_{0L}, Z_{0R}, H_1 and θ_L; z_i is the z coordinate of the entrance point of the incident ions. Several predictions, using this surface topography, were made for the $^{18}O(p,\alpha_0)^{15}N$ reaction at E_p=1.78 MeV and 165° for ^{18}O step concentration profiles for thick flat and non-flat Fe_3O_4 oxides. The differential cross section parameterization used in these simulations was available as a computer file (4).

4. RESULTS AND DISCUSSION

In the predictions for nuclear reactions we used available data of stopping power of the required ions in the samples (11-14).

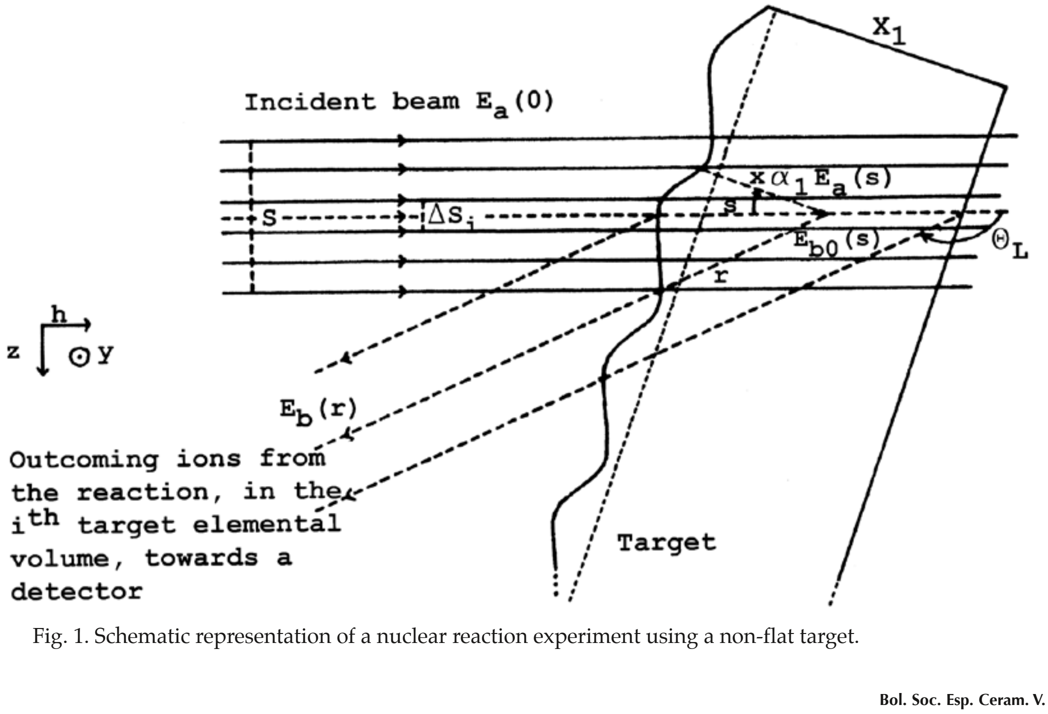

Fig. 1. Schematic representation of a nuclear reaction experiment using a non-flat target.

4.1. $^{12}C(d,p_0)^{13}C$ reaction

In simulating proton spectra of the ^{12}C (d, p_0) ^{13}C reaction for flat target we used our measured values of the differential cross-section shown in Fig. 3 for E_d : 0.790- 2.060 MeV, at 165° and 135° (5). These values were complemented with other published data: at 160° in the range E_d: 0.390- 0.790 MeV (15); at 135° in the range E_d: 0.500- 0.780 MeV (16), multiplied by a factor of 0.88, to join our data starting at 0.790 MeV. Several simulations were made for perpendicular incidence of the deuteron beam, at E_d=1.40 MeV and 165°. Lindhard-Scharff energy straggling theory and Fermi theory for multiple scattering were used. The computer simulations for a thick flat target, whose stages are shown in Fig. 4, show that final yield resembles the differential cross section curve.

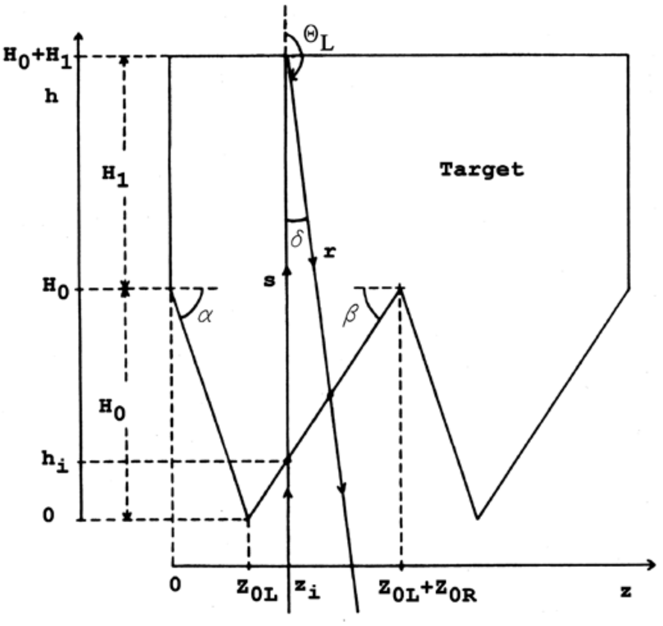

Fig. 2. Parameterization of a target having an asymmetric triangular surface contour.

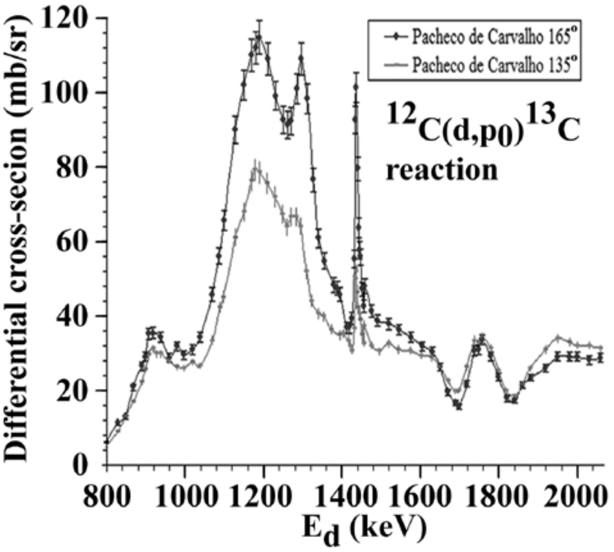

Fig. 3. Differential cross section measurements for the $^{12}C(d,p_0)^{13}C$ reaction at laboratory angles of 165° and 135° (5).

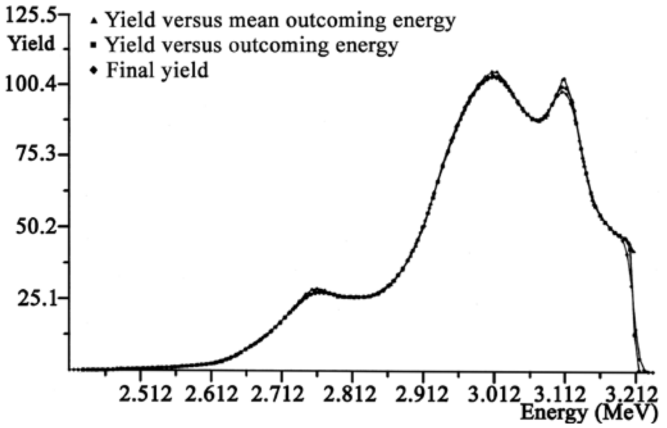

Fig. 4. Stages of the predictions for the $^{12}C(d,p_0)^{13}C$ reaction at E_d=1.40 MeV and 165°, for a ^{12}C target thickness X_1= 10.000 μm.

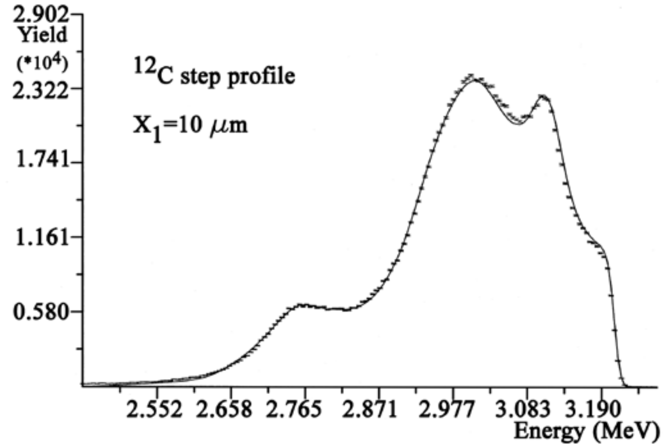

Fig. 5. Computed fit to the spectrum of the $^{12}C(d,p_0)^{13}C$ reaction in the *S1* C target, at E_d=1.40 MeV and 165°.

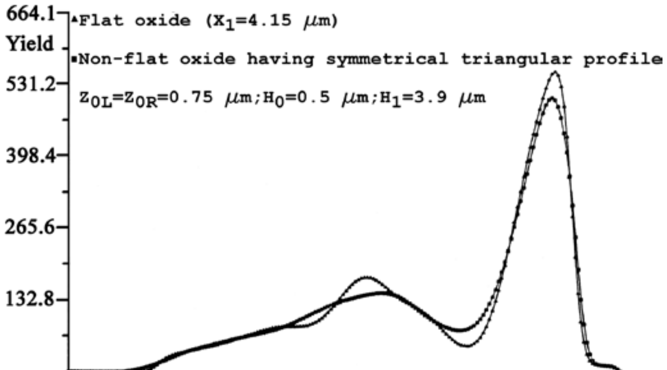

Fig. 6. Predicted spectra of the $^{18}O(p,\alpha_0)^{15}N$ reaction in flat and non-flat oxides with symmetric triangular profile, at E_p=1.78 MeV and 165°.

Good computer fits were obtained to the spectra of the S1 pyrolitic graphite target at various energies of the deuteron beam, both at 165º and 135º. Uniform concentration profiles of ^{12}C were used along considerable depths near the surface of the target. The results are illustrated through Fig. 5 at E_d = 1.40 MeV and 165º.

4.2. $^{18}O(p,\alpha_0)^{15}N$ reaction

Simulations for the $^{18}O(p,\alpha_0)^{15}N$ reaction at E_p=1.78 MeV and 165º for ^{18}O step concentration profiles, for thick flat and non-flat Fe_3O_4, oxides have shown (Figs. 6-7) that pronounced surface topography significantly affect the shapes of the energy spectra.

The oxidized steel S2 sample containing ^{18}O, was analysed through 1.78 MeV proton beams, an energy slightly above the 1.763 MeV resonance, at normal incidence and θ_L =165º. The corresponding spectrum of the $^{18}O(p,\alpha_0)^{15}N$ reaction is shown in Fig. 8. The predictions used available differential cross section data for this reaction (17). A good computed fit to data was obtained. An ^{18}O oxide was found, with uniform concentration and X_1=4.4 μm thickness. This thickness is higher than the value determined from the "resonance method" of analysis using the 1.763 MeV resonance, as the method of the present work permits higher depth resolution.

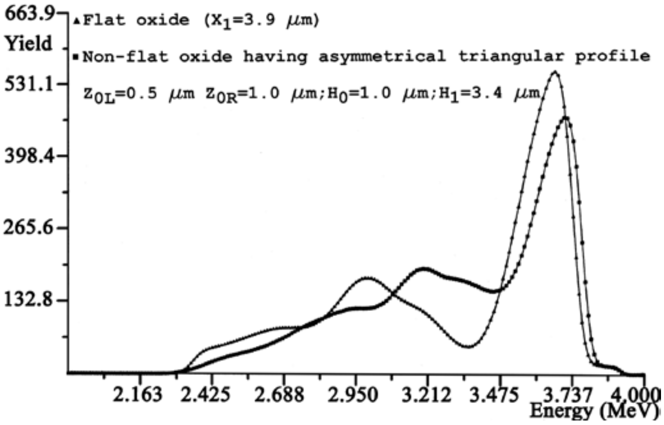

Fig. 7. Predicted spectra of the $^{18}O(p,\alpha_0)^{15}N$ reaction in flat and non-flat oxides with asymmetric triangular profile, at E_p=1.78 MeV and 165º.

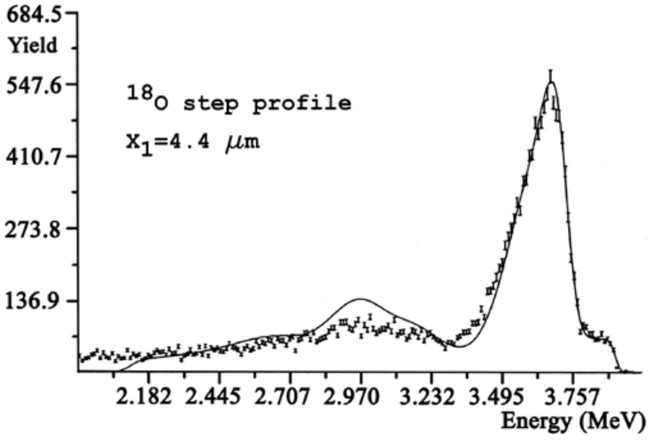

Fig. 8. Computed fit to data of the $^{18}O(p,\alpha_0)^{15}N$ reaction in the *S2* target, for E_p=1.78 MeV and 165º.

5. CONCLUSIONS

This work has shown successful applications of surface analysis by nuclear reactions, for depth profiling of ^{12}C and ^{18}O nuclei. Computer simulation permitted predictions to be made both for flat targets and for non-flat targets having asymmetric triangular surface contours, showing surface topography effects on energy spectra. The spectral predictions resulted in good descriptions of experimental spectra obtained for thick samples and considerable depths close to the surface. The presented results would be very difficult to obtain by non-nuclear techniques.

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